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Research Paper

Mechanisms Involved in Osmotic Backwashing of Fouled Forward Osmosis (FO) Membranes

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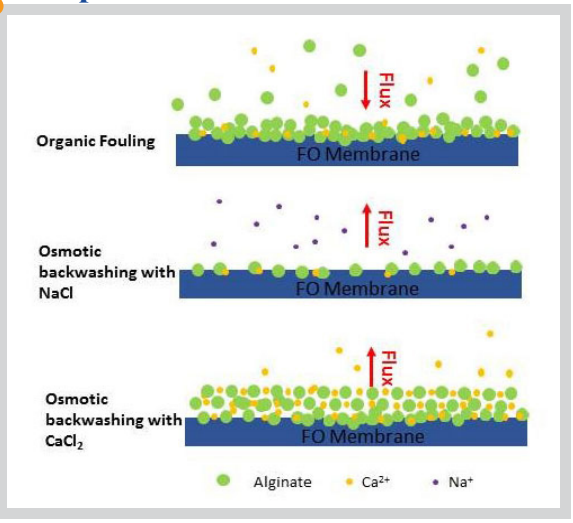
Highlights

- Osmotic backwashing efficiently cleans most alginic acid fouled FO membranes
- Fouling removal and 100% PWF recovery were obtained for all Ca concentrations tested
- Limitations in efficiency arise with Ca present in the osmotic backwashing solution
- Limitations in efficiency arise with a 4 M NaCl DS solution during fouling
- PWF is not a good indicator for cleaning efficiency

Graphical abstract



Abstract

Organic matter leads to one of the biggest problems in membranes: fouling. Developing efficient cleaning processes is therefore crucial. This study systematically examines how alginic acid fouling formed under different physical and chemical conditions affect osmotic backwashing cleaning efficiency in forward osmosis (FO). The fouling layer thickness before and after osmotic backwashing was measured by confocal laser scanning microscopy in order to assess cleaning efficiency, along with pure water flux (PWF) measurements. Osmotic backwashing was found to be very efficient. In the absence of Ca^{2+} in the feed solution, the alginate fouling thickness was <33 µm. The presence of 2.5 mM Ca^{2+} in the feed solution promoted the formation of a compact fouling layer, with a thickness of 173 µm. One minute of backwashing using 0.7 M NaCl, fully restored the PWF and reduced the fouling layer thickness down to <6 µm. However, backwashing with less than 0.7 M NaCl was less effective, with 26 µm of fouling remaining, despite a complete PWF recovery. Backwashing also became less effective when the initial membrane fouling flux increased using a draw solution (DS) of 4 M NaCl, with 91 µm of fouling remaining, despite a full PWF restoration. The use of Ca^{2+} in the osmotic backwashing DS caused the fouling layer to expand and not be removed due to flux reversal and the interaction between the alginic acid layer and Ca^{2+}. A reduction in the PWF recovery was obtained, showing the type of salt used for backwashing has a severe influence on cleaning efficiency.

1. Introduction

The process of forward osmosis (FO) has recently been the focus of much interest in wastewater reclamation, with reported encouraging results for many different types of wastewater [1-7]. In forward osmosis, two liquids with different osmotic pressures are separated by a semi permeable membrane. The difference in osmotic pressure between the two solutions acts as a driving force for water to permeate from the feed solution (FS) side of low osmotic pressure to the draw solution (DS) side of high osmotic pressure. Gebreyohannes et al. [2], for example, successfully used FO to dehydrate olive mill wastewater. The waste volume reduced by 71% using a DS of 3.7 M $MgCl_2$, and the olive mill waste was rejected by 98%. Hickenbottom et al. [4] recovered 80% of clean water from contaminated drilling wastewater using a FO process, where 99% rejection of dissolved organic carbon was

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achieved.

FO has the potential for many low energy applications, which include indirect desalination where wastewater reclamation is coupled with reverse osmosis (RO) desalination. In indirect desalination, the FO membrane is used to permeate clean water from the wastewater FS side to the seawater DS side. This process uses "free osmotic energy" due to the difference in osmolarity between the DS and FS sides, leading to a diluted seawater DS, which can be subsequently further desalinated by a low-pressure RO step [8-11].

However, the presence of organic matter in wastewater can lead to fouling, which causes a reduction in membrane flux, hence productivity. Several studies have shown that FO membrane performance can be severely affected by fouling through flux reduction with time [2, 4, 12, 13]. Mi and Elimelech [14, 15] reported a 24 hour decrease in FO membrane flux up to 60% due to the formation of a loose, but thick organic fouling layer on the membrane surface. Despite being loose, however, up to 80% of the flux decline during FO fouling was found to be irreversible [13], and 100% flux recovery was not achieved even when using chemical cleaning on FO membranes treating secondary wastewater effluent [16, 17].

Furthermore, fouling has also been shown to affect rejection by RO and FO membranes due to cake enhanced concentration polarization [18], as well as due to properties of the contaminants, of the membranes and of the foulants [19]. Xie et al. [20] showed that trace contaminant rejection decreased from 85% to 60% for fouled FO membranes. Research into effective cleaning methods for organic fouling removal from FO membranes is therefore crucial for the successful operation and maintenance of FO processes.

Currently, both chemical and chemical free cleaning methods have been investigated for membrane processes. Chemical methods include cleaning with chlorine, alkaline, acid and detergent solutions. However, chemical cleaning can damage the membrane, hence affecting its performance and longevity. Membrane cleaning through chlorination has been carried out for FO [16] and NF/RO [18] membranes, but it was found to compromise the integrity of the membranes' active layer, affecting their permeability and solute rejection, including trace contaminant rejection. Wang et al. [21] obtained an increase in water flux and solute flux when cleaning FO membranes with NaOH, SDS and/or Alconox, where the latter caused a solute flux increase by a factor of 3. Similar outcomes have been obtained for NF membranes cleaned with NaOH, where contaminant rejection was reduced [20]. On the other hand, Valladares et al. [16] increased fouling reversibility up to 93.6% in FO membranes used to treat secondary wastewater effluent by using a mixture of Alconox and EDTA. However, these chemicals have been found to be toxic to aquatic life and ecosystems [22], hence presenting an environmental issue. Alternatives to chemical cleaning should hence be researched.

Physical cleaning can be carried out by using air scouring, by increasing the crossflow velocity and/or by flushing the membrane with deionised water. Valladares et al. [16] examined natural organic matter fouling of membranes and restored 89.5% of the initial membrane flux using air scouring. Motsa et al. [23] used membrane surface flushing at increased crossflow velocities with ultrapure water, obtaining a 98%, 93% and 91% flux restoration for three consecutive fouling and cleaning cycles. Cleaning efficiency was lost, as the fouling layer became more difficult to remove after three cycles. Kim et al. [13] and Mi and Elimelech [15], on the other hand, fully restored the pure water flux (PWF) after 1 hour and 15 min, respectively, when cleaning FO membranes fouled with alginic acid and different Ca^{2+}concentrations by surface flushing at increased cross-flow velocities (from 8 $cm.s^{-1}$ to 34 $cm.s^{-1}$ and 8.5 $cm.s^{-1}$ to 21 $cm.s^{-1}$, respectively). However, cleaning efficiency is only reported in terms of flux recovery and no surface imaging was carried out in any of the above reported studies. As clearly shown by Motsa et al., despite a high flux recovery in the first cycle, the reduction in flux recovery for subsequent cycles suggests fouling was present on the membrane, hence flux recovery might be an insufficient measurement for assessing cleaning efficiency. Boo et al. [24] showed that fouling conditions with silica nanoparticles actually affected cleaning efficiency. In this study, they increased the crossflow velocity from 8.5 to 25.6 $cm.s^{-1}$, which recovered 95% of the membrane flux lost in FO. However, the cleaning method efficiency was reduced when the fouling FS pH was increased from 4 to 9, as only 80% of the flux restoration was achieved. At higher pH, the silica nanoparticles were destabilised, resulting in increased particle aggregation. This resulted in the formation of a thicker and less porous fouling cake layer which was more difficult to remove. This shows cleaning efficiency is highly dependent on the fouling feed parameters, and hence on the fouling layer characteristics. Understanding fouling characteristics through imaging is therefore crucial when understanding cleaning efficiency in membrane processes. Furthermore, not only do some of these studies show an incomplete cleaning efficiency in terms of flux recovery, but the cleaning strategies presented would be energetically costly (e.g. air scouring and increased cross-flow for >15 min) and require process downtime, as well as for some cases using the product for cleaning, i.e. permeated water.

Osmotic backwashing is a novel cleaning method which involves reversing the permeated flux of water through the membrane, with the aim to physically remove fouling. This is achieved in RO by introducing a high salinity pulse on the feed side, for example, hence drawing water from the permeate side to the feed side by osmosis. Several studies have shown promising results for osmotic backwashing cleaning of RO membranes [25, 26]. Ramon et al. [27] restored 93% to 97% of the flux of a fouled RO membrane after osmotic backwashing and Tow et al. [28] restored 80% of the initial flux of a RO membrane fouled with alginic acid and 1 mM $CaCl_2$ by osmotic backwashing and higher crossflow velocity.

FO requires no hydraulic pressure to produce flux, and therefore it has lower irreversible fouling tendency compared to RO [29]. This indicates greater potential for cleaning through osmotic backwashing, achieved in FO by simply swapping the high osmotic pressure DS with the low osmotic pressure FS (or using clean water as FS), thus reversing the driving force for permeation across the membrane, resulting in flux reversal. An advantage of osmotic backwashing cleaning over other methods previously mentioned are: (1) not requiring the use of chemicals, (2) having the potential to take less time and hence have a lower process downtime, (3) not requiring an increase in cross-flow velocity or air scouring for an efficient removal of the fouling layer, hence not recurring on extra energy costs.

However, as with RO, process parameters including the chemistry of the fouling solution, as well as membrane flux, affect the rate and extent of fouling in FO [14]. These influences are in turn expected to affect cleaning efficiency, as per the study by Boo et al. In fact, studies on the efficiency of osmotic backwashing cleaning of FO membranes have produced a wide range of results: these have spanned from unsuccessful [16, 30] to highly effective [4, 23, 31], with initial flux recoveries ranging from 0%, [30] to 100% [4]. Arkhangelsky et al. [30] applied osmotic backwashing in hollow fibre FO membranes fouled with 3.88 $g.L^{-1}$ $CaCl_2$, 2.84 $g.L^{-1}$ Na_2SO_4, 1.11 $g.L^{-1}$ NaCl and saturated with 130% $CaSO_4$. They concluded that osmotic backwashing in FO was ineffective, with 0% flux restored. In contrast, Hickenbottom et al. [4] recovered 100% of the flux and visually removed the fouling layer from the FO membrane surface using osmotic backwashing, when processing drilling wastewater from a shale gas field as a FS, and NaCl solution as a DS. Motsa et al. [23] used osmotic backwashing to clean organic fouling on FO membranes where divalent cations (Mg^{2+} and Ca^{2+}) were present: the NaCl DS of varying concentrations was replaced with ultrapure water, whilst the FS remained unchanged, comprising of a solution of sodium alginate, Mg^{2+} and Ca^{2+}. Like Hickenbottom et al. [4], osmotic backwashing restored 100%, 99% and 93% of the flux after three consecutive fouling and cleaning cycles. The influence of osmotic backwashing duration was studied by Blandin et al. [32]. FO membranes fouled with humic acid and alginic acid were osmotically backwashed and flux recoveries of 60%, 85% and almost 100% for osmotic backwashing durations of 15 minutes, 1 hour and 1 hour with high cross flow velocity were achieved, respectively. This shows the influence that osmotic backwashing parameters also have on the cleaning efficiency of FO membranes.

The wide differences obtained in initial flux restoration in FO after osmotic backwashing is due to several factors, including fouling conditions and fouling layer characteristics, as well as osmotic backwashing conditions. The lack of understanding of the fundamental mechanisms involved hence invite a study of the parameters affecting fouling characteristics, and consequently, osmotic backwashing efficiency. As previously mentioned, limited to no information is provided in the literature on fouling layer properties before and after osmotic backwashing, hence imaging techniques were systematically used in this study to assess fouling layer removal efficiency. This study systematically demonstrates how different fouling conditions in FO using alginic acid, calcium and different process parameters, including osmotic pressure difference during fouling, affect osmotic backwashing efficiency. Osmotic backwashing cleaning was also carried out under different conditions of osmotic backwashing DS concentration. Furthermore, since alginic acid and calcium are known to interact and complex [33, 34], the type of salts used in the osmotic backwashing DS, namely NaCl and $CaCl_2$ were also studied.

2. Materials and methods

2.1. FO membrane: aquaporin inside™

A commercial aquaporin based FO membrane was used for all experiments (Aquaporin Inside™, Denmark). The membrane has a thin film composite (TFC) structure, with a polyamide active layer embedded with aquaporin proteins, as well as support layers made of polyester and non-woven polyester. The thickness of the membrane is approximately 4 to 5 mm

thick. The membrane was stored dry. Before use, the membrane was gently washed and stored in ultrapure water (Avidity, UK) overnight at 4°C. During this study, a second batch of Aquaporin Inside™ FO membranes was used, which had different properties compared to the previous batch. It was noted that flux values obtained under identical conditions were higher for the second batch of membranes. Results obtained with the second batch of experiments are identified in the relevant Figure captions (*i.e.* Figures 3, 6 and 7). These membranes were stored wet before use. The membrane was then rinsed and cut to fit the membrane cell before use.

2.2. Organic fouling feed solution

The organic fouling feed solution consisted of alginic acid sodium salt (AA) derived from brown algae (Sigma Aldrich, UK). AA was chosen as a model foulant for polysaccharides found in wastewater [35], as well as its suitability for use with confocal imaging microscopy. The fouling solutions were made from a stock of AA concentration of 2 $g.L^{-1}$. The stock solution was stirred overnight to ensure the AA was fully dissolved. For each experiment, a feed solution of 200 $mg.L^{-1}$ AA and a background electrolyte solution of 1 mM $NaHCO_3$ (Sigma-Aldrich, UK), 20 mM NaCl (Fisher Scientific, UK) and 2.5 mM $CaCl_2$ (Sigma-Aldrich, UK) was prepared, unless otherwise specified: the solution was stirred overnight to ensure complete mixing. This fouling solution was chosen in order to accelerate and promote irreversible fouling [13-15].

2.3. Forward osmosis crossflow system

The FO membrane cell was a custom-built Perspex membrane cell. The effective membrane area was of 0.0048 m^2 (membrane width of 25 mm and length of 191 mm). The membrane was placed between two channels, each of 3 mm in height. The membrane cell was arranged in co-current flow in order to decrease the stress on the membrane in the cell [4, 8, 14].

Figure 1 illustrates the FO crossflow system used in this study. Two gear pumps of variable speeds (Cole Parmer, Vernon Hills, IL) delivered a constant flow of 1 $L.min^{-1}$ to each of the channels of the membrane cell. A system of valves was put in place in order to select the reservoirs which feed the two channels of the membrane cell: either (1) the Feed Solution FS_f and Draw Solution DS_f reservoirs were used for fouling or (2) the Osmotic Backwashing Feed Solution FS_{obw} and Osmotic Backwashing Draw Solution DS_{obw} reservoirs were used for osmotic backwashing. The flux through the membrane during fouling or osmotic backwashing was obtained by recording the weight changes of the DS_f and DS_{obw} reservoirs, respectively, which were placed on a balance (Ohaus, US), with the weight logged at known intervals throughout the experiment. The temperature in the FO system was maintained at 20 ± 1°C by a cooling bath (Haake F3, UK), through coils submerged in both FS_f and DS_f tanks.

Both lines feeding the membrane cell were fitted with flowmeters (104 Flo-Sen McMillan, UK) and pressure transducers (PX219-30V45G5V, Omega, UK) to monitor flow conditions. Each tank contained one thermocouple (TJ2-CPSS-M6OU-200-SB, Omega, UK) and one conductivity meter (Cond 340i meter, WTW, Germany) in order to monitor temperature and quantify reverse salt flux, respectively. The temperature, pressure and flowrate were logged with a DAQ 54 Omega data logger (Omega, UK). The weight of the reservoirs were logged using LabVIEW. The entire rig was set up on a heavy marble slab to reduce any vibrations.

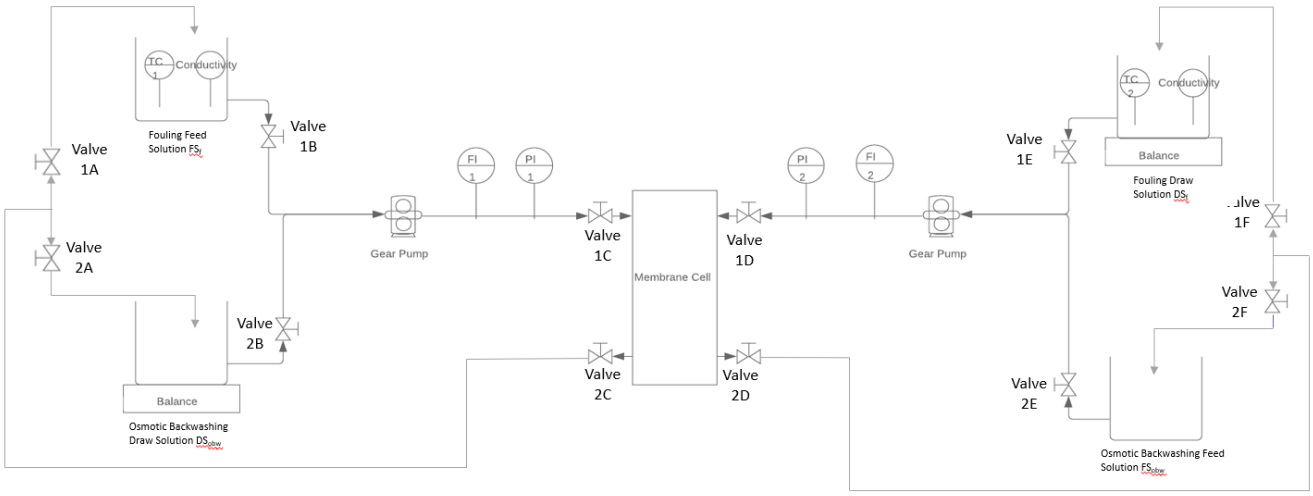

Fig. 1. P&ID of the FO crossflow system.

2.4. Fouling protocol

The membrane was carefully taped in the membrane cell using double sided tape to ensure no movement inside the cell occurred when manipulating it. The PWF of the membrane was tested for 30 minutes with a Feed Solution FS_f of ultrapure water and a Draw Solution DS_f of 0.7 M NaCl, used to simulate seawater [6, 8, 36]. This was followed by membrane fouling, where the solution in the Feed Solution FS_f tank was changed to the chosen fouling solution, and the solution in the Draw Solution DS_f tank was changed to the chosen draw solution, in most cases to 0.7 M NaCl, unless otherwise specified. The fouling experiments were stopped when 300 mL of water permeated from the Feed Solution FS_f tank to the Draw Solution DS_f tank. This translated to fouling experiments lasting around 20 hours for a DS_f of 0.7 M NaCl and around 4 hours for a DS_f of 4 M NaCl. See Fig. **2.** (A) for an illustration of the fouling process.

2.5. Cleaning Protocol

Cleaning by osmotic backwashing was carried out immediately after fouling. The flow from the gear pumps was firstly stopped, followed by valve manipulation, *i.e.* closing valves 1A, 1B, 1E, 1F and opening valves 2A, 2B, 2E, 2F (Fig. **1**.), in order to switch from the Feed Solution FS_f and Draw Solution DS_f tanks used for fouling to the Osmotic Backwashing Feed Solution FS_{obw} and Osmotic Backwashing Draw Solution DS_{obw} tanks to be used for osmotic backwashing. The FS_{obw} and DS_{obw} solutions used were ultrapure water and a high salt concentration solution, respectively. In most cases, a DS_{obw} of 0.7 M NaCl was used, but other NaCl concentrations were tested as well, as specified. A DS_{obw} solution of 0.5 M $CaCl_2$ was also used, in order to study the effect of Ca^{2+} on the DS_{obw}. The flow was restarted, osmotic backwashing was carried out for 1 minute at 1 $L.min^{-1}$ crossflow rate, and the flux was measured by weighing the DS_{obw} tank on a balance (Ohaus, US). See Fig. **2.** (B) for an illustration of the osmotic backwashing cleaning process.

For comparison purposes, cleaning by surface flushing was carried out for the study of the influence of Ca^{2+} concentration in the FS_f. To implement this, both the FS_{obw} and DS_{obw} consisted of deionised water, to ensure there was no transmembrane flux. Surface flushing was carried out for 1 minute at 1 $L.min^{-1}$.

After cleaning, the flow was stopped, the valves were manipulated again, and the FS_f and DS_f solutions were substituted by the same solutions as those used for the initial PWF measurements described in section 2.4, in order to

assess PWF recovery after osmotic backwashing. The membrane was then carefully removed from the membrane cell in order to carry out microscopy visualisation techniques, as described in the next section. The FO crossflow system was then cleaned, by firstly rinsing with ultrapure water, followed by a 30 minute recirculation of 0.1 M NaOH. Then, 0.1 M HCl was added in order to neutralise the NaOH solution. The rig was thoroughly rinsed again with deionised water until the conductivity was 0 $\mu S.cm^{-1}$ and pH was neutral, measured with a pH probe (VWR, Germany).

Five consecutive cycles of fouling and cleaning experiments were also performed to examine osmotic backwashing cleaning efficiency for several cycles. The FS_f solution used was 200 $mg.L^{-1}$AA, 1 mM $NaHCO_3$, 20 mM NaCl and 2.5 mM $CaCl_2$. Two DS_f, namely 0.7 M NaCl and 4 M NaCl, were tested for fouling durations of 22 hours and 4 hours, respectively. Fouling was followed by 1 minute of cleaning with an osmotic backwashing DS_{obw} of 0.7 M NaCl. At the end of the 5 cycles, the PWF was measured.

2.6. Sample staining

In order to understand osmotic backwashing efficiency, the concanavalin A (Con A) dye was used to stain a fouled membrane and a fouled membrane followed by osmotic backwashing. This study used Con A Alexa Fluor™ 488 Conjugate (ThermoFisher Scientific), displaying bright green fluorescence. The lectin Con A binds to mannose residues of glycoproteins and is a widely used dye to stain polysaccharides like alginic acid. Stock solutions of 1 $mg.ml^{-1}$ of Con A were prepared and stored in the freezer in 0.1 ml volumes.

At least 3 membrane samples were cut from the middle of the membrane, with a size of 20 by 20 mm. These were next positioned on a glass slide, and using a micropipette, 50 µl of thawed Con A solution was applied directly on the fouling layer (or the osmotic backwashed fouling layer). Next, these samples were incubated for 30 minutes in the dark. Double sided tape was used to make an adapted washer, which was placed around the sample to act as an O-ring, followed by a glass coverslip which was positioned on top of the O-ring. The O-ring, positioned between the glass slide and the coverslip, was used to stop the coverslip from compressing the gel fouling layer.

2.7. Confocal laser scanning microscopy (CLSM)

A Zeiss LSM 880 with Fast Airyscan CLSM, and with a laser scanning module fitted on an inverted microscope (Zeiss) and an argon laser, was used to image each membrane sample in 3 different areas. Images were recorded with an excitation wavelength of 488 nm and generated by obtaining Z-stacks of the fouling layer. This microscope has lateral resolution of 200 nm for 2D and 3D Z stacks and 500 to 600 nm axial resolution for Z-stacks. The lowest increment quantified by the microscope is < 25 nm. Next, Imaris™ software was used for image analysis, in order to precisely visualize and measure the fouling layer thickness. The heterogeneity of the fouling layer was not taken into account in this study, since it was found that the surface coverage of the membrane by the fouling layer remained even. The membrane had some background fluorescence, hence its thickness was determined based on several measurements and was found to be 19.91±4.2 µm. This thickness was subtracted from the total fouling layer thicknesses determined by the confocal microscope.

2.8. Total organic carbon analysis

The concentrations of TOC in the FS_f and DS_f were measured in order to determine by mass balance how much AA deposited on the membrane surface during fouling. For that purpose, a Total Organic Carbon analyser (TOC-V CPH) was used in Non Purgeable Organic Carbon mode (NPOC) (Shimadzu, Milton Keyes, UK). Samples of 10 ml were taken from the FS_f and DS_f at the start and end of the fouling experiments and at least 4 more samples were taken throughout the experiment. Next, 2 M HCl was added to the samples and they sparged for 1.5 minutes with N_2 in the TOC in order to remove inorganic carbon. A 10 $mg.L^{-1}$ standard of potassium hydrogen phthalate (PHP) was used throughout the TOC measurements to ensure results were accurate.

3. Results and discussion

3.1. Impact of FS chemistry on FO fouling and osmotic backwashing

Ca^{2+} ions are known to form complexes with carboxylic groups present in alginic acid and form a thick, compact fouling layer on the membrane surface [14, 25]. Varying Ca^{2+} concentration in the fouling FS_f will hence affect the alginic acid fouling layer characteristics [37]. The extent to which FS_f chemistry affects osmotic backwashing cleaning efficiency was assessed for the first time to the authors' knowledge using confocal microscopy imaging before and after cleaning, as well as through PWF recovery. The membranes were fouled using alginic acid with varying concentrations of $CaCl_2$ in the FS_f. Osmotic backwashing with DS_{obw} of different NaCl concentrations, namely 0.7 M and 4 M, were applied to the fouled FO membranes. Figure 3 shows examples of representative confocal images of fouled and 0.7 M NaCl osmotic backwashed FO membranes, used to characterise the fouling layer before and after cleaning. For comparison purposes, membrane surface flushing was also applied, since it showed to be efficient in some FO cleaning studies [15].

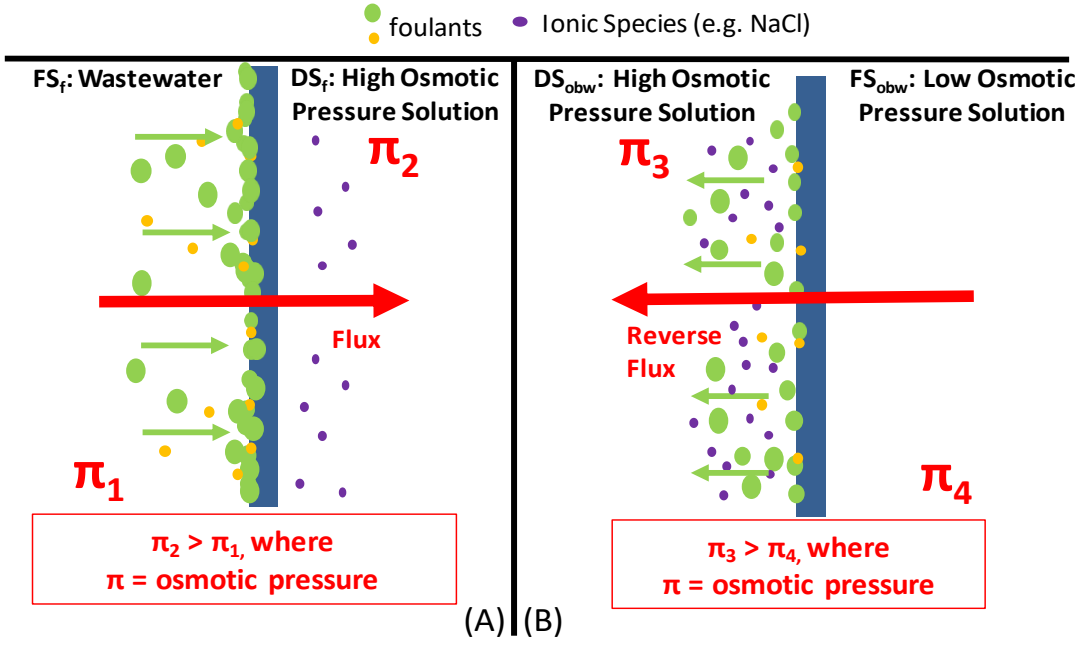

Fig. 2. Illustration of the FO (A) fouling and (B) osmotic backwashing cleaning procedure. During fouling, a wastewater FS_f of low osmotic pressure (π_1) is circulated on the active layer side of the FO membrane and a high osmotic pressure DS_f (π_2), such as seawater, is circulated on the support layer side. During osmotic backwashing cleaning, the wastewater FS_f is substituted by a high osmotic pressure DS_{obw} (π_3) solution, such as seawater, and the DS_f is substituted by a low osmotic pressure FS_{obw} (π_4) solution, such as pure water. This causes flux reversal during cleaning.

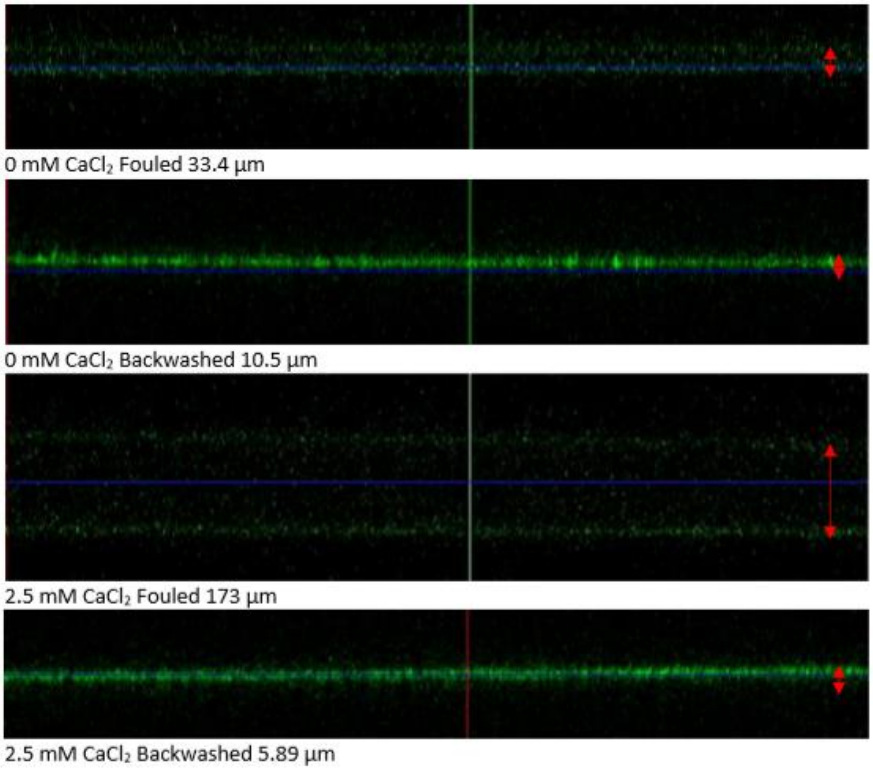

Fig. 3. Confocal images of fouled and osmotic backwashed FO membranes (FS_f: 200 mg.L^{-1} AA, 20 mM NaCl and 1 mM $NaHCO_3$ with varying Ca^{2+} concentrations; DS_f: 0.7 M NaCl; FS_{obw}: pure water; DS_{obw}: 0.7 M NaCl).

As shown in Figure 4, the fouling layer thicknesses ranged between 25 and 33 µm when Ca^{2+} in the FS_f ranged between 0 and 1 mM. For higher Ca^{2+} concentrations of 1.5 and 2.5 mM, the fouling layer thickness increased substantially, ranging between 168-174 µm: Ca^{2+} ions form complexes with the carboxylic groups present in AA, neutralizing the AA negative charge, causing a thick gel layer to form, which resulted in fouling of the membrane [15, 33].

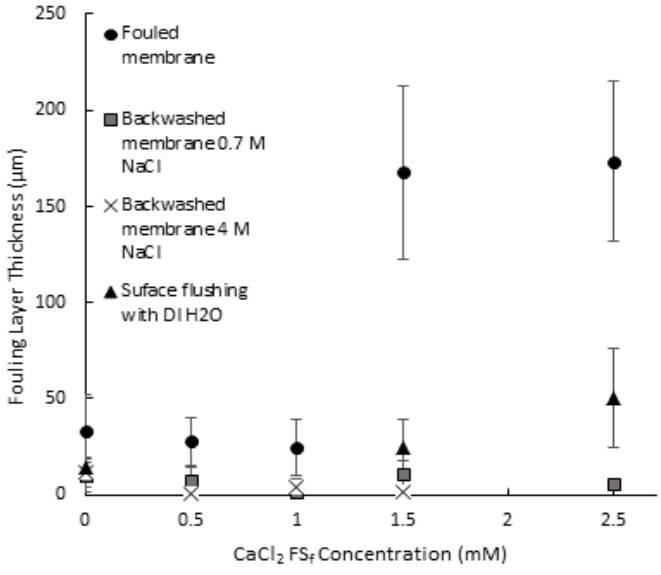

Fig. 4. Fouling layer thickness before and after osmotic backwashing from confocal microscopy images, as a function of FS_f Ca^{2+} concentration during fouling (fouling conditions: DS_f = 0.7 M NaCl; FS_f = 200 mg.L^{-1} AA, 20 mM NaCl, 1 mM $NaHCO_3$, varying concentrations of $CaCl_2$, duration = 18 hours; osmotic backwashing conditions: DS_{obw} = 0.7 M or 4 M NaCl, FS_{obw} = deionised water, duration = 1 minute; surface flushing conditions: FS_{obw} and DS_{obw} = deionized water, duration = 1 minute, crossflow rate: 1 L.min^{-1}; Error bars show standard deviation of repeated experiments).

Interestingly, the $CaCl_2$ concentration in FS_f had a small effect on the flux reduction in the present study: 43% of the initial flux was reduced in the presence of 0 mM $CaCl_2$, as opposed to 41% reduction in the presence of 2.5 mM $CaCl_2$, even though the fouling layer produced was thicker in the latter case. A similar result was obtained by Parida and Ng [38] and Kim et al. [13], where the absence and presence of Ca^{2+} did not substantially affect FO performance in terms of flux decline. In FO, AA fouling layers have been depicted as loosely formed and "fluffier" compared to those in RO, which are denser and more compact [15, 29]. As such, the thicker fouling layer formed in FO for 2.5 mM $CaCl_2$ in the FS_f combined with a DS_f of 0.7 M NaCl, offered no added resistance to flux. Motsa et al. [23] found that the deposition of AA onto the FO membrane resulted in a loose and less compact fouling layer, which allowed for water permeation.

Surface flushing has been previously reported as effective in cleaning fouled FO membranes, including alginic acid fouling. Motsa et al. [23] showed that 15 minutes of surface flushing at a higher cross-flow velocity with deionised water could restore 98% of the PWF. In the present study, surface flushing for 1 minute with deionised water with a constant cross-flow velocity showed a reduced cleaning efficiency with increased $CaCl_2$ concentration in the FS_f (Figure 4). A fouling layer of 14.6 µm thickness stayed on the membrane surface for experiments carried out without Ca^{2+}. The fouling thickness that remained on the membrane surface for Ca^{2+} concentrations of 1.5 mM and 2.5 mM, increased to 25 µm and 50 µm, respectively. The PWF restoration reduced from 97% for 0 mM Ca^{2+}, to 83% for 1.5 mM, and 76% for 2.5 mM Ca^{2+}. Lee and Elimelech [37] determined that adhesion forces for AA fouling in RO membranes increased with increasing Ca^{2+} concentration, hence explaining why surface flushing became less efficient for higher Ca^{2+} concentrations in the FS_f. Kim et al. [13] and Mi and Elimelech [15], on the other hand, fully restored the PWF after 1 hour and 15 min, respectively, of surface flushing of alginic acid fouled FO membranes with different concentrations of Ca^{2+}. The differences could be due to time used for surface flushing, i.e. 1 minute in the present study vs 1 hour or 15 min in the studies mentioned above, coupled with increasing cross-flow velocities in these studies during surface flushing (from 8 cm.s^{-1} to 34 cm.s^{-1} and 8.5 cm.s^{-1} to 21 cm.s^{-1}, respectively). The reason why 1 minute and the same cross-flow velocity of 1 L.min^{-1} were tested in the present study was to compare with osmotic backwashing efficiency, which will be described subsequently. Surface flushing was hence deemed inadequate, especially at higher Ca^{2+} concentrations in the FS_f (Figure 4).

The effect of the osmotic backwashing DS_{obw} concentration was tested

for 0.7 M and 4 M NaCl. Average osmotic backwashing fluxes of 8.3±2.9 $L.m^{-2}.h^{-1}$ and 26.4±3.7 $L.m^{-2}.h^{-1}$ for 0.7 M and 4 M NaCl were obtained, respectively. As shown in Figure 4, both NaCl concentrations used in the DS_{obw} succeeded in eliminating the fouling layer from the membrane surface for all FS_f Ca^{2+} concentrations, reducing them to less than 11.5 µm, as well as restoring the flux to 99.9±5.9% and 113.1±12.8%, respectively. The efficiency of osmotic backwashing cleaning is hence superior compared to surface flushing for 1 minute. Note that flux recoveries > 100% have been reported in the literature [26], potentially due to AA fouling remaining on the membrane surface, which would make the membrane more hydrophilic [26, 39]. Osmotic backwashing is more efficient than surface flushing because it uses permeate drag perpendicular to the membrane surface, which removes the fouling layer by shear force. Furthermore, during osmotic backwashing, ion exchange occurs between the ions in the fouling layer and the ions in the DS_{obw}, causing structural variations to the fouling layer. These physical changes cause the fouling layer to weaken, making its removal from the membrane surface much easier [26]. Furthermore, osmotic backwashing for 1 min and a DS_{obw} of 0.7 M NaCl was as, or more efficient, in removing the fouling layer and fully restoring the PWF when compared to surface flushing for >15 min at much higher cross-flow velocities [13, 15, 23], showing advantages with lower chemical free cleaning time and lower energy consumption.

In the studies by Valladares-Linares et al. [6, 16], osmotic backwashing efficiently removed the synthetic wastewater foulants from the FO membranes in a plate and frame assembly, but PWF recovery was non-existent due to internal concentration polarization (ICP) caused by the accumulation of ionic species inside the FO membrane. However, the osmotic backwashing method applied consisted of soaking the membrane for 12 h in a 4% NaCl solution, while deionized water was recirculated inside the plate and frame lumen space of the cell. Arkhangelsky et al. [30] obtained a negative PWF recovery for similar reasons, despite having cross-flow. Osmotic backwashing was inefficient for these studies due to the adopted protocol, which promoted ICP, whilst it was highly efficient for the present study. Hence, cleaning by osmotic backwashing is more efficient for shorter cleaning times and for cross-flow FO systems, where ICP is avoided.

The fouling layer thickness increase for higher Ca^{2+} concentrations in the FS_f was found not to offer added resistance to osmotic backwashing flux. When the Ca^{2+} concentration in the FS_f increased from 1 to 1.5 mM, the fouling layer thickness increased from 25 µm to 168 µm. The resulting osmotic backwashing fluxes were however similar (5.24 $L.m^{-2}.h^{-1}$ and 6.25 $L.m^{-2}.h^{-1}$, respectively). This could be due to the fact that in FO, the fouling layer formed, as previously mentioned, is "fluffier" and less compact, therefore not offering added resistance to osmotic backwashing flux, in the same way no added resistance was obtained for the flux during fouling [15].

Since a DS_{obw} of 0.7 M NaCl provided similar results to a DS_{obw} of 4 M NaCl (Figure 4), DS_{obw} of lower NaCl concentrations were tested for optimization purposes. This is shown in Figure 5.

The lower the DS_{obw} concentration, the lower the efficiency in removing the fouling layer: the fouling layers left after osmotic backwashing had thicknesses of 6 µm for 0.7 M NaCl, 18 µm for 0.1 M NaCl and 26 µm for 0.05 M NaCl. The osmotic backwashing fluxes were 13.5, 11.56 and 10.05 $L.m^{-2}.h^{-1}$ for the 0.7, 0.1 and 0.05 M NaCl solutions, respectively (please note these osmotic backwashing fluxes are higher due to the usage of a second batch of FO membranes, as mentioned in section 2.1). DS_{obw} concentrations < 0.7 M NaCl hence do not offer sufficient drag force to remove the fouling layer from the membrane surface. The higher osmotic backwashing flux obtained with the 0.7 M NaCl DS_{obw} results in a drag force which is sufficient to overcome the forces of adhesion between the AA foulant layer and the surface of the membrane, and therefore allowing for the removal of the fouling layer. A minimum of 0.7 M NaCl DS_{obw} solution is therefore necessary to remove the fouling layer for the conditions tested in this study (Figure 5). Furthermore, as previously mentioned, the occurrence of ion exchange between the fouling layer salts and the DS_{obw} also occurs, which causes a break-up of the AA-Ca^{2+} bonds and weakening of the fouling layer, making it easier to remove [26]. A minimum concentration required for the DS_{obw} solution could explain why osmotic backwashing is not always 100% efficient in the literature [1, 12], as the osmotic backwashing process was not optimized and an insufficient reversed permeate drag force was generated to fully remove the fouling layer.

Restoration of PWF was high for all cases studied in Figure 5, with a recovery of 103.5±3.7%. However, despite a full PWF recovery, there is still fouling on the surface, as seen in Figure 5. This fouling can accumulate during subsequent fouling and cleaning cycles, which will affect membrane performance. Relying on PWF restoration measurements is hence insufficient to assess cleaning efficiency, and imaging techniques should hence be used as well.

A 0.7 M NaCl DS_{obw} was determined as the optimal solution to clean the membrane for the studied conditions. Other salt solutions, such as Ca^{2+}, can also be used to create the same osmotic pressure difference between both sides of the membrane, hence the same osmotic backwashing flux. However, Ca^{2+} is known to interact with alginic acid, as previously described, hence osmotic backwashing with $CaCl_2$ might affect the cleaning efficiency.

3.2. Impact of DS chemistry on FO osmotic backwashing

In order to assess if the presence of Ca^{2+} in DS_{obw} affects cleaning efficiency, an osmotic backwashing DS_{obw} of 0.5 M $CaCl_2$, giving a similar osmotic backwashing flux as 0.7 M NaCl was tested. The osmotic backwashing fluxes obtained were 8.3±2.9 $L.m^{-2}.h^{-1}$ for 0.7 M NaCl DS_{obw} and 10.1±0.9 $L.m^{-2}.h^{-1}$ for 0.5 M $CaCl_2$ DS_{obw}. The results are shown in Figure 6.

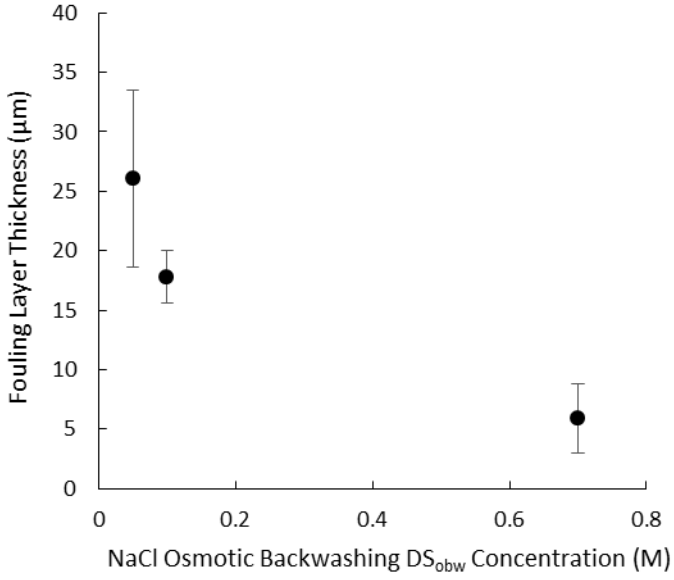

Fig. 5. Fouling layer thickness after osmotic backwashing from confocal microscopy images, as a function of DS_{obw} NaCl concentration (fouling conditions: DS_f = 0.7 M NaCl; FS_f = 200 $mg.L^{-1}$ AA, 20 mM NaCl, 1 mM $NaHCO_3$, 2.5 mM $CaCl_2$, duration = 18 hours; osmotic backwashing conditions: duration = 1 minute; Error bars show standard deviation of repeated experiments. Experiments were carried out using a new batch of membranes)

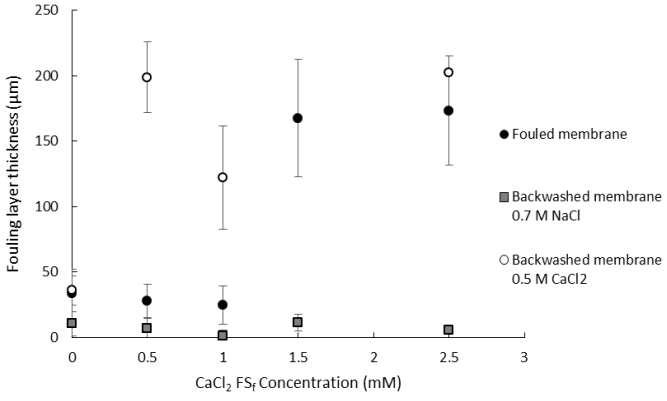

Fig. 6. Fouling layer thickness before and after osmotic backwashing from confocal microscopy images, as a function of the Ca^{2+} in the FS_f, where 0.7 M NaCl or 0.5 M $CaCl_2$ were used as osmotic backwashing solutions (fouling conditions: DS_f = 0.7 M NaCl; FS_f = 200 $mg.L^{-1}$ AA, 20 mM NaCl, 1 mM $NaHCO_3$, duration = 18 hours; osmotic backwashing conditions: duration = 1 minute; The error bars display the standard deviation of repeated experiments).

When comparing cleaning with 0.7 M NaCl versus 0.5 M $CaCl_2$ DS_{obw}, it is clear the latter is ineffective in removing the fouling layer, despite a similar osmotic backwashing flux obtained with 0.7 M NaCl DS_{obw}, which is able to remove the fouling layer (Figure 6). In fact, as shown in Figure 6, 0.5 M $CaCl_2$ DS_{obw} causes the fouling layer thickness to increase, when compared to the thickness of the fouled layer before osmotic backwashing. The fouling layer thicknesses increased by 171 µm for 0.5 mM $CaCl_2$, by 97 µm for 1 mM $CaCl_2$ and by 29 µm for 2.5 mM $CaCl_2$, after cleaning with 0.5 M $CaCl_2$ DS_{obw}. A possible explanation is that the osmotic backwashing flux causes the fouling layer to expand, but the carboxylic groups of the expanded AA layer bond with the Ca^{2+} ions in the DS_{obw} and the adhesion forces between AA molecules increase, making their removal more difficult [14, 37]. This increase in adhesion forces between AA molecules could not be overcome by the perpendicular shear force caused by the osmotic backwashing flux, and hence the fouling layer was not removed.

Osmotic backwashing with $CaCl_2$ was also inefficient at restoring the PWF. Flux restorations of 100%, 83%, 91% and 77% were achieved for FS_f of 0 mM, 0.5 mM, 1 mM and 2.5 mM $CaCl_2$, respectively, and a DS_{obw} of 0.5 M $CaCl_2$. This was opposed to PWF restorations of 99.9±5.9% for DS_{obw} of 0.7 M NaCl. A DS_{obw} solution with Ca^{2+} ions is hence not suitable to clean a membrane by osmotic backwashing, since the Ca^{2+} ions interact with the AA fouling layer, even if osmotic backwashing only takes place for 1 minute. The superior cleaning efficiency provided by NaCl DS_{obw}, confirms that monovalent ions are more suitable for cleaning AA fouled FO membranes compared to osmotic backwashing with $CaCl_2$ DS_{obw}.

The study by Gebreyohannes et al. [2] treated pectin rich olive mill wastewater with FO, and subsequently applied osmotic backwashing with a DS_{obw} of 3.7 M $MgCl_2$. Much like alginic acid, pectin, a polysaccharide, is also known to interact with divalent ions such as Ca^{2+} and Mg^{2+} and form a gel. However, in the presence of Mg^{2+}, pectin forms gels with a more open microstructure and less intertwined network when compared to Ca^{2+} [40]. This can potentially explain why the PWF recovery for Gebreyohannes et al. [2] after osmotic backwashing with Mg^{2+} was of 95%, higher than the PWF recovery from the present study. A DS_{obw} solution of Mg^{2+} could hence be researched for the removal of more challenging alginic acid fouling, as it might have a lower interaction with the fouling layer but allow for a higher osmotic pressure difference to be applied during cleaning, hence increasing the reversal flux.

3.3. Impact of operational flux on FO fouling and osmotic backwashing

The initial membrane flux during fouling was increased by increasing the DS_f concentration, in order to assess the impact of flux on fouling and osmotic backwashing efficiency. Membrane flux has been shown to affect fouling characteristics.

Xie et al. [20] compared initial fluxes of 9 and 20 $L.m^{-2}.h^{-1}$ for humic acid and colloidal FO fouling using SEM and found that the higher initial flux resulted in a more compact layer. Assessing how flux impacts fouling, and therefore how it indirectly affects cleaning efficiency, is required. Results are presented in Figure 7.

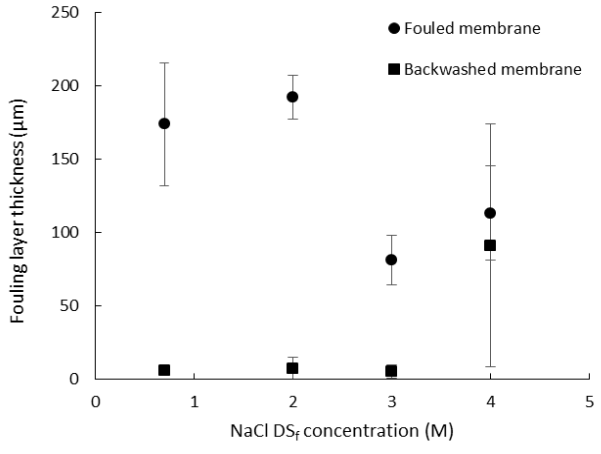

Fig. 7. Fouling layer thickness before and after osmotic backwashing from confocal microscopy images, as a function of DS_f NaCl concentration (fouling conditions: FS_f = 200 $mg.L^{-1}$ AA, 20 mM NaCl, 1 mM $NaHCO_3$, 2.5 mM $CaCl_2$, duration = varied; osmotic backwashing conditions: DS_{obw} = 0.7 M NaCl, FS_{obw} = deionised water, duration = 1 minute; Error bars show standard deviation of repeated experiments).

Increasing the NaCl DS_f concentration from 0.7 M to 4 M, increases the initial membrane flux from 5.87±0.46 $L.m^{-2}.h^{-1}$ to 21.95±3.75 $L.m^{-2}.h^{-1}$. As a consequence, the fouling layer thickness reduces from > 173 µm down to < 113 µm for DS_f concentrations above 3 M NaCl (Figure 7). Xie et al. [41] showed that drag forces through the membrane caused compaction of the AA layer, explaining the results shown in Figure 7: the fouling layer becomes thinner and denser. TOC analysis of the FS_f proves this further, as the TOC accumulated on the membrane was 36.3 $mgC.L^{-1}$ and 34.4 $mgC.L^{-1}$ for a DS_f solution of 0.7 M NaCl and 4 M NaCl, respectively. This confirms further that the layer resulting from 4 M NaCl DS_f has a higher density because it is 60 µm thinner despite having a similar amount of TOC as the 0.7 M NaCl DS_f.

The reverse salt flux was significantly affected by the DS_f concentration, increasing from 12 $g.m^{-2}.h^{-1}$ to 26 $g.m^{-2}.h^{-1}$ for a DS_f of 0.7 M NaCl and 4 M NaCl, respectively. This is expected, since a higher transmembrane osmotic pressure difference increases the driving force for reverse flux of the salt. This is important, since a previous study [42] determined that in the presence of Ca^{2+} ions, an AA layer thickness increased from 25 nm to >150 nm when a monovalent salt concentration, namely KCl, increased from 0 mM to 100 mM KCl. However, the same study found that when the KCl concentration was further increased to 300 mM, the opposite happened, and the AA fouling layer reduced from 150 nm to 50 nm. They explain that this is the alginate layer's response to changes in salt concentration at high ionic strength and highlights the instability of the alginate layer. The same phenomena is observed here: the reverse salt flux of Na^+ ions from the DS_f into the FS_f, increased the FS_f ionic strength, and possibly resulted in a reduction of the fouling layer thickness (Figure 7).

Similarly with RO, a higher rate of flux decline will occur in FO when the initial flux is increased [30]. In the present study, the FO flux decline increased from 10% for an initial flux of 6 $L.m^{-2}.h^{-1}$ (i.e. 0.7 M NaCl DS_f) to 32% for an initial flux of 21 $L.m^{-2}.h^{-1}$ (i.e. 4 M NaCl DS_f). This can be due to, as with RO, a higher initial flux offering higher drag force perpendicular to the membrane surface, which results in a fouling layer that is denser (Figure 7) and displays a higher resistance to flux. However, unlike in RO, increasing the DS_f concentration results in an increase in ICP, which reduces the osmotic pressure difference and hence, also reduces the flux [43, 44].

The flux restorations achieved after cleaning with 0.7 M NaCl DS_{obw} were 103%, 113%, 97% and 104% for DS_f of 0.7, 2, 3 and 4 M NaCl, respectively. For the 0.7 M NaCl DS_f, 96.5% of the fouling was removed, with a remaining fouling thickness of less than 6 µm. Increasing the DS_f concentrations further up to 3 M NaCl, resulted in less than 6.6 µm of fouling remaining on the membrane surface after osmotic backwashing cleaning. Cleaning with 0.7 M NaCl DS_{obw} is hence effective for all DS_f tested (Figure 7), even for the case of a denser and more compact fouling layer obtained for a 3M NaCl DS_f: osmotic backwashing is hence effective in removing the fouling layer and in fully restoring the flux (Figure 7).

There was, however, an exception: osmotic backwashing the fouling layer formed with a 4 M NaCl DS_f, where 91 µm of the fouling layer remained on the surface. For this particular case, the fouling layer was not efficiently removed when using a DS_{obw} of 0.7 M NaCl, as it was dense and compact, and the adhesion forces between foulant molecules were too high to be overcome by osmotic backwashing. The results by Motsa et al. [23] suggest a similar cause for cleaning inefficiency, where a 6 M NaCl DS_f was used during fouling and a DS_{obw} of 1.8 M NaCl was used during osmotic backwashing, with PWF recoveries for several subsequent cycles varying between 84 and 93%. A higher DS_{obw} concentration or longer osmotic backwashing duration could potentially completely remove the fouling layer formed with a 4 M NaCl DS_f. For all DS_f concentrations studied, the flux was fully restored, including a DS_f of 4 M NaCl, where 91 µm of fouling remained on the membrane surface. This confirms, once more, that a full PWF recovery does not translate into complete removal of fouling, showing the importance of carrying out surface imaging to verify the true competence of membrane cleaning techniques.

3.4. Fouling and cleaning cycles

Despite the PWF being recovered in most studied cases, and only a low quantity of fouling was not removed from the membrane surface by osmotic backwashing (Figure 7), the fouling that remains could potentially affect fouling adhesion and flux decline for subsequent cycles of fouling and cleaning. Adhesion forces have been shown to be proportional to fouling rate in a previous study [37], hence even a small number of adhesion sites on the membrane surface being occupied by organic matter molecules can increase membrane fouling and cause subsequent cleaning efficiency to reduce [15]. Hence, five consecutive fouling and osmotic backwashing cleaning cycles were performed, in order to assess if the fouling that remained on the membrane surface after a first cleaning cycle affects fouling and osmotic

backwashing cleaning in FO further. The membrane was fouled for 22 hours with a 0.7 M NaCl DS_f and a 200 mg/L AA, 1 mM $NaHCO_3$, 20 mM NaCl and 2.5 mM $CaCl_2$ FS_f for each cycle, followed by 1 minute of osmotic backwashing with 0.7 M NaCl DS_{obw}, as this was determined to be the optimum osmotic backwashing solution (Figure 5). The PWF was measured after 5 cycles were completed.

As can be seen in Figure 8, the membrane flux was reduced from approximately 16 $L.m^{-2}.h^{-1}$ to 4 $L.m^{-2}.h^{-1}$ for every fouling cycle. After carrying out osmotic backwashing with 0.7 M NaCl DS_{obw} for 1 minute, the flux is restored to 16 $L.m^{-2}.h^{-1}$ for every cycle. The reverse salt flux stayed constant for every fouling cycle, increasing from 18 $g.m^{-2}.h^{-1}$ for the first cycle, to 20 $g.m^{-2}.h^{-1}$ for the fifth cycle. The fouling behaviour hence remains consistent throughout each cycle, showing that osmotic backwashing is effective.

The osmotic backwashing flux decreased from cycle 3 to cycle 5, 33.8±5.9 $L.m^{-2}.h^{-1}$ to 19.4±1.1 $L.m^{-2}.h^{-1}$, respectively, with the layer potentially becoming more resistant to osmotic backwashing after numerous cycles. However, this osmotic backwashing flux is still enough to remove most of the fouling layer after 5 cycles, which had a thickness of 3.07±1.97 µm, as well as obtaining a high PWF restoration, which decreased from 103 to 97%. The loose fouling gel layer formed on the membrane surface in the FO process [15] allows for its efficient removal using osmotic backwashing. This is similar to results reported by Motsa et al. [23] and Martinetti et al. [45], where osmotic backwashing restored 100%, 99% and 93% of the flux and 93% and 83% of the flux for consecutive fouling and cleaning cycles, respectively. Holloway et al. [31] used FO membranes to concentrate anaerobic digester centrate. Fouling was carried out for two 20 hour cycles before 10 minutes and 20 minutes osmotic backwashing with a 50 $g.L^{-1}$ NaCl solution was performed. This restored approximately 85% and less than 90% of the initial flux, respectively. These results indicate that the driving force for osmotic backwashing might need to increase in order to efficiently clean the FO membrane.

To further test the efficiency of osmotic backwashing cleaning, more challenging fouling conditions were adopted by implementing a higher fouling flux using a 4 M NaCl DS_f during five consecutive cycles. This allowed assessing how the remaining 91 µm of fouling layer after the first cycle (Figure 7) affected fouling and cleaning further.

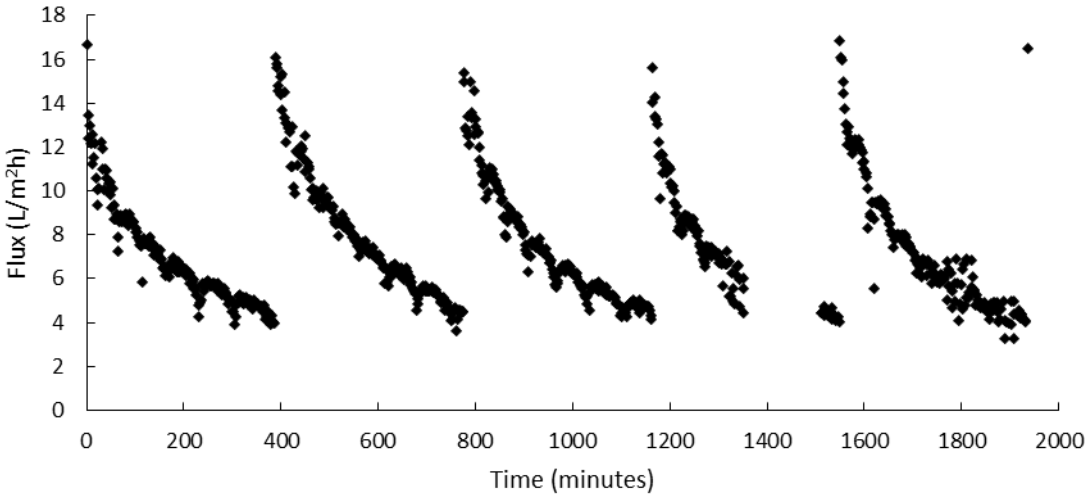

Fig. 8. Membrane flux for five successive rounds of fouling and osmotic backwashing cleaning (fouling conditions: DS_f = 0.7 M NaCl, FS_f = 200 mg/L AA, 20 mM NaCl, 1 mM $NaHCO_3$, 2.5 mM $CaCl_2$, duration = 22 hours; osmotic backwashing conditions: DS_{obw} = 0.7 M NaCl, FS_{obw} = deionised water, duration = 1 minute. These experiments were carried out with a new batch of membranes).

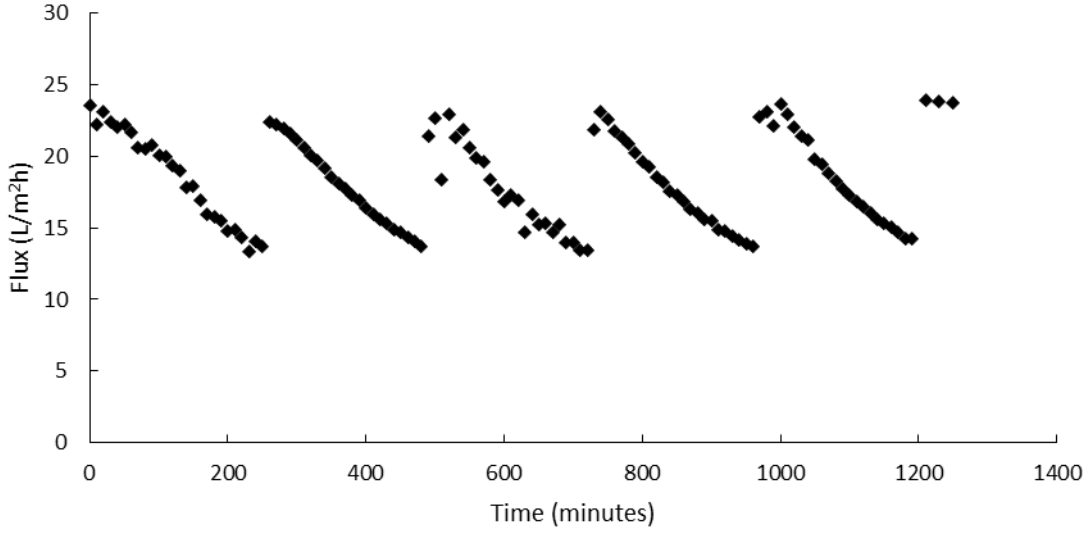

Fig. 9. Membrane flux for five successive rounds of fouling and osmotic backwashing cleaning (fouling conditions: DS_f = 4 M NaCl, FS_f = 200 mg/L AA, 20 mM NaCl, 1 mM $NaHCO_3$, 2.5 mM $CaCl_2$, duration = 4 hours; osmotic backwashing conditions: DS_{obw} = 0.7 M NaCl, FS_{obw} = deionised water, duration = 1 minute. These experiments were carried out with a new batch of membranes).

The results in Figure 9 show a similar outcome to those obtained for a lower fouling flux using 0.7 M NaCl DS_f (Figure 8). For every fouling cycle, which lasted 4 hours, the flux decreased from approximately 23 $L.m^{-2}.h^{-1}$ to 14 $L.m^{-2}.h^{-1}$, followed by a full flux restoration after 1 minute osmotic backwashing cleaning with 0.7 M NaCl DS_{obw}. The similarity in membrane flux reduction for every cycle, shows that the fouling remaining on the membrane surface after each cycle had no effect on the flux, even after 4 successive fouling experiments. At the end of the fouling cycles, 6.19±3.92 µm of fouling remained on the membrane surface, showing an efficient removal with osmotic backwashing.

TOC analysis of the FS_f further showed that the fouling rate was consistent between different cycles since the amount of foulant accumulated on the membrane surface did not change with repeated cycles: 37.5 $mgC.L^{-1}$ and 37.4 $mgC.L^{-1}$ were obtained for cycle 1 and cycle 5, respectively. The same consistency was obtained for the measured reverse salt flux for a membrane fouled with a DS_f of 4 M NaCl: 26 $g.m^{-2}.h^{-1}$ was obtained for the first cycle, reducing to 22.2±1.4 $g.m^{-2}.h^{-1}$ for subsequent cycles. The osmotic backwashing flux was also relatively unaffected throughout the several fouling cycles, decreasing from 18 $L.m^{-2}.h^{-1}$ to 16 $L.m^{-2}.h^{-1}$ from cycle 1 to cycle 5, respectively. Maintaining a constant osmotic backwashing flux after each fouling cycle is important to ensure a high enough permeate drag is obtained in order to efficiently remove the fouling layer that forms after each fouling cycle.

4. Conclusions

Osmotic backwashing was shown to be a very efficient cleaning technique for organically fouled FO membranes. Once the cleaning method was optimized, it was shown to be efficient even under challenging fouling conditions like the ones tested in this study, as well as when subjected to several fouling and cleaning cycles. As the Ca^{2+} concentration in the FS increases, the fouling layer becomes more difficult to remove via surface flushing, indicating higher adhesion forces of the fouling layer with increasing Ca^{2+} concentration. Osmotic backwashing is, however, an efficient cleaning method. This is because the fouling layer formed in FO is soft and loosely formed when compared with RO, making it easier to remove when reversing the flux direction through the membrane. Osmotic backwashing with 0.7 M NaCl for just 1 minute, efficiently restored the flux and removed the fouling layer from the membrane surface when the increase of Ca^{2+} concentration in the FS_f increased the fouling layer thickness and cohesion.

However, the chemical composition of the osmotic backwashing DS_{obw} was shown to play a crucial role in osmotic backwashing cleaning efficiency. The presence of Ca^{2+} in the osmotic backwashing DS_{obw} can severely reduce cleaning efficiency compared to a DS of NaCl for the same osmotic backwashing flux, as Ca^{2+} interact with the fouling layer and do not allow for its removal. Other limitations in osmotic backwashing of FO membranes were found when high fouling fluxes were used, i.e. when the DS_f was increased to 4 M NaCl, since this resulted in a more compact and denser fouling layer. However, despite a poor removal rate of the fouling layer, where 91 µm were left on the membrane, 100% of the PWF was restored. Flux measurements alone are not sufficient to prove that the cleaning method employed was efficient, and imaging techniques are hence required.

In order to assess the osmotic backwashing cleaning method for FO membranes, multiple consecutive fouling and osmotic backwashing cycles were performed. In this study, the flux was successfully restored after 5 cycles using DS_f concentrations of 0.7 and 4 M NaCl. The resulting fouling layer was also successfully removed down to less than 6.19±3.92 µm. These results show the efficiency of the cleaning method adopted, even for challenging fouling conditions, eliminating the need for cleaning chemicals and reducing cleaning downtime. Since the fouling layer in FO is formed in the absence of applied pressure, and is therefore loose and less dense, its fouling effects are easier to reverse. The same will not occur in RO, as shown by Xie et al. [41], hence studying osmotic backwashing in fouled RO membranes and understanding the fundamental mechanisms involved is required, as these are expected to be different from FO, since there is no salt reverse flux and no ICP effects as well.

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